

# Analytical approximation of biodegradation rate for in situ bioremediation of groundwater under ideal radial flow conditions

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## Abstract

We derive the long-term biodegradation rate of an organic contaminant (substrate) for an in situ bioremediation model with axisymmetric flow conditions. The model presumes that a nonsorbing electron acceptor is injected into a saturated homogeneous porous medium which initially contains a sorbing substrate and attached indigenous microorganisms. The derived analytical removal rate depends upon the injection flow rate, the initial substrate and supplied acceptor concentrations, the stoichiometric coefficient for acceptor utilization, and the sorption characteristics of the substrate; the removal rate does not depend upon the dispersion parameters, microbial kinetic parameters, and initial biomass concentration. Numerical simulations confirm the analytical results. The insensitivity of the long-term removal rate to the microbial kinetic parameters and initial biomass concentration suggests that precise estimation of these data may not be necessary to assess bioremediation effectiveness. In the numerical results, however, there exists an initial transient phase during which the removal rate depends upon microbial growth kinetics. This initial phase is significantly prolonged if the initial substrate and injected acceptor concentrations are at nutrient-limiting levels, or if the microbial kinetic parameters and initial biomass concentration do not yield efficient microbial growth and substrate utilization. © 1998 Elsevier Science B.V.

**Keywords:** In situ bioremediation; Transport model; Radial flow; Analytical biodegradation rate

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## 1. Introduction

Contamination of soil and groundwater by hazardous substances has been widely recognized as an important environmental and health problem that must be urgently solved. The Resource Conservation and Recovery Act (RCRA) lists more than 37,000 contaminated sites that are required for remediation in the United States (Abelson, 1992). In situ bioremediation is an emerging technology which is considered more cost-effective and time-saving than conventional remediation techniques such as excavation of contaminated soil and pump-and-treat methods (Alexander, 1991; Lee et al., 1988; Shevah and Waldman, 1995). A recent trend of in situ bioremediation has headed toward the treatment of more recalcitrant organic contaminants like halogenated compounds (Brown et al., 1993; McCarty and Semprini, 1993).

Despite tremendous efforts towards technological innovations based upon microbiological and geochemical investigations, field applications of in situ bioremediation often show decontamination that is less effective than what is expected from laboratory experiments (Sturman et al., 1995). Successful engineered in situ bioremediation relies on the efficient formation of a subsurface bioreactor, a so-called biologically active zone (BAZ) (Alvarez-Cohen, 1993). Good mixing between an organic pollutant (substrate) and an electron acceptor is required for effective formation of a BAZ in which microbial activities are stimulated. Poor mixing due to aquifer heterogeneities is one possible explanation for low in situ biodegradation rates (MacQuarrie and Sudicky, 1990; Schäfer and Kinzelbach, 1992).

Odenrantz (1992) and Odenrantz et al. (1993) described another important mixing process related to substrate sorption for an idealized one-dimensional Cartesian bioremediation model in which a nonsorbing electron acceptor is supplied to the inlet boundary of a homogeneous soil column which uniformly contains the substrate and indigenous microorganisms. Injection of the electron acceptor displaces the substrate and produces a leading concentration front of the acceptor downstream and a trailing concentration front of the substrate upstream. The substrate migrates downstream at a retarded velocity due to its sorption, while the acceptor migrates at the pore water velocity. This velocity difference causes the two fronts to overlap one other (advective mixing) and to form a mixing zone in which biodegradation occurs.

In numerical simulations of the one-dimensional Cartesian bioremediation model, Odenrantz (1992) and Odenrantz et al. (1993) observed travelling waves for both solute fronts, that is, the fronts migrate downstream with constant mean velocity and profile shape. Oya and Valocchi (1997) presented more detailed analysis of this interesting front behaviour and concluded that travelling waves in the bioremediation model result from the interactions between the advective mixing and biodegradation processes. Using the mathematical properties of a travelling wave (Auchmuty et al., 1986; Britton, 1986, pp. 61–71; Ortoleva and Schmidt, 1985; Volpert et al., 1994), Oya and Valocchi (1997) theoretically derived the long-term substrate removal rate. The resulting analytical removal rate is independent of the dispersion parameters, the initial biomass concentration, and the microbial kinetic parameters. The removal rate is instead dependent upon the initial substrate and supplied acceptor concentrations, the sorption parameter of the substrate (retardation factor), the stoichiometric coefficient for acceptor

consumption, and the groundwater flow velocity. Although the analytical investigation was restricted to simplified one-dimensional bioremediation conditions, these findings are significant and useful for understanding the fundamental characteristics of fate and transport of the biologically reactive solutes.

Another simplified bioremediation system that has practical relevance is a case with radial flow since input of electron acceptors and other limiting nutrients is often accomplished via injection wells. This type of in situ bioremediation problem may be represented by a one-dimensional axisymmetric (radial flow) transport-reaction model, if the aquifer is assumed homogeneous with negligible natural-gradient flow and if the water table rise due to the injection is insignificant relative to the saturated thickness for an unconfined aquifer. Because the ideal radial flow field is nonuniform, it is significantly more complex mathematically than one-dimensional Cartesian flow and there have been no analytical solutions for radial flow biodegradation systems reported in the literature. Under the radial flow condition, the fronts do not produce travelling waves because the front velocity decreases with radial distance, and therefore we cannot apply the travelling wave theory to the radial flow system. The primary objectives of this paper are thus to find an alternative analytical method to derive the substrate removal rate under this idealized axisymmetric flow condition, to examine its validity by comparing with numerical results, and to understand the relationship of the biodegradation characteristics to those for the Cartesian model.

## 2. Governing equations

The one-dimensional axisymmetric in situ bioremediation model has the coordinate  $r$  with an origin at the center of the injection well and presumes uniform constituent concentrations in the angular direction. Fig. 1 shows the idealized bioremediation scenario discussed in this paper. The water table rise near the injection well is assumed negligible compared with the saturated thickness of the aquifer (Fig. 1a). Fig. 1b shows the initial and boundary conditions of the substrate  $S$ , electron acceptor  $A$ , and biomass  $M$ . The aquifer initially has uniform distributions of the substrate and biomass with respective concentrations of  $S_0$  and  $M_0$ . The injected solution contains only the electron acceptor at a concentration of  $A_0$ .

As often observed in the field, it is assumed in the model that the organic substrate sorbs onto aquifer solids (e.g., Schwarzenbach and Westall, 1981) while the acceptor (e.g., oxygen or nitrate) does not (e.g., Chen et al., 1992). Although kinetic sorption/desorption is an important factor for bioavailability of nutrients (Bouwer et al., 1994; Criddle et al., 1991; Fry and Istok, 1994), we assume linear equilibrium sorption because this study focuses on finding an analytical method for estimating the substrate removal rate without using the travelling wave theory. Although recent research has addressed the transport of microorganisms (e.g., Harvey and Barber, 1992; Hornberger et al., 1992), in this study the active microorganisms are assumed immobile (attached biomass) and to be able to utilize only aqueous-phase compounds for growth (Mihelcic et al., 1993; Ogram et al., 1985). The initial biomass concentration is assumed to be at a steady background level  $M_0$  which results from an equilibrium state between cell

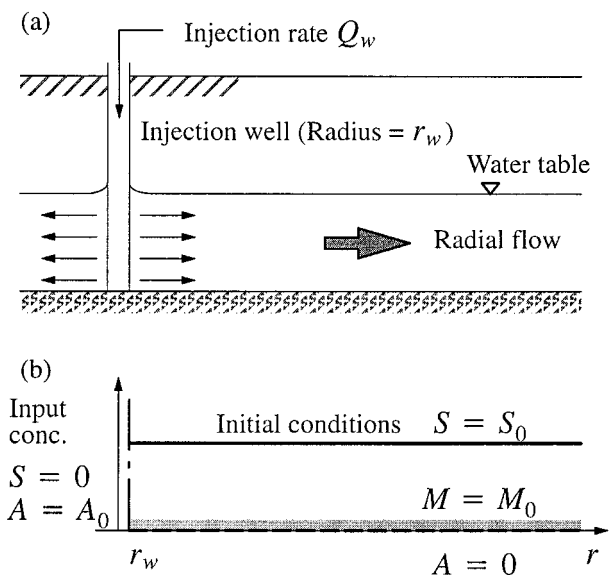


Fig. 1. Schematic illustration of (a) the aquifer geometry under the hypothetical remediation scenario and (b) initial and boundary conditions of all constituents on the radial coordinate  $r$ .

growth on naturally-occurring substrates and endogenous cell decay (Chiang et al., 1991). For further simplicity, it is assumed that there is no acceptor consumption by endogenous cell decay and other indigenous microbes, that the hydrodynamic conditions do not change due to cell growth and decay, and that the diffusion properties of the solutes are identical.

These assumptions lead to the following equations for the axisymmetric model:

$$R_d \frac{\partial S}{\partial t} - \frac{1}{r} \frac{\partial}{\partial r} \left( rD \frac{\partial S}{\partial r} \right) + v \frac{\partial S}{\partial r} = -q_m f_{BD} \quad (1)$$

$$\frac{\partial A}{\partial t} - \frac{1}{r} \frac{\partial}{\partial r} \left( rD \frac{\partial A}{\partial r} \right) + v \frac{\partial A}{\partial r} = -F q_m f_{BD} \quad (2)$$

$$\frac{\partial M}{\partial t} = Y q_m f_{BD} - d(M - M_0) \quad (3)$$

$$f_{BD} = M \left( \frac{S}{K_S + S} \right) \left( \frac{A}{K_A + A} \right) \quad (4)$$

where  $t$  is the time [T],  $R_d$  is the retardation factor of the substrate,  $D$  is the hydrodynamic dispersion coefficient [ $L^2 T^{-1}$ ],  $v$  is the pore water velocity [ $LT^{-1}$ ] in the radial direction,  $q_m$  is the maximum utilization rate of the substrate [ $M_S M_M^{-1} T^{-1}$ ],  $F$  is the stoichiometric coefficient for the electron acceptor consumption [ $M_A M_S^{-1}$ ],  $Y$  is the cell yield coefficient [ $M_M M_S^{-1}$ ], and  $d$  is the cell decay rate constant [ $T^{-1}$ ]. The dependent variables are the concentrations of the substrate  $S$  [ $L^{-3} M_S$ ], electron acceptor

$A$  [ $L^{-3}M_A$ ], and biomass  $M$  [ $L^{-3}M_M$ ]. Eq. (4) is the nonlinear component of the multiplicative Monod function for dual limitation to microbial uptake of the solutes. In the equation,  $K_S$  and  $K_A$  are the half-saturation constants of the substrate [ $L^{-3}M_S$ ] and the electron acceptor [ $L^{-3}M_A$ ], respectively. Note that the biomass concentration  $M$  is on a pore volume basis. The dispersion coefficient  $D$  consists of molecular diffusion,  $D_m$  [ $L^2T^{-1}$ ], and mechanical dispersion,  $\alpha_L v$ , in which  $\alpha_L$  is the longitudinal dispersivity [ $L$ ], such that (Bear, 1972)

$$D = \alpha_L v + D_m \quad (5)$$

Note that the pore water velocity  $v$  varies with  $r$  according to the assumption of ideal radial flow; that is,  $v = Q_w / (2\pi\theta r)$ , where  $Q_w$  is the injection rate per unit thickness of the aquifer [ $L^2T^{-1}$ ], and  $\theta$  is the porosity.

According to Fig. 1b, the model Eqs. (1)–(3) are to be solved subject to the inlet boundary and initial conditions (see Fig. 1b)

$$\begin{aligned} \left( -rD \frac{\partial S}{\partial r} + rvS \right)_{r=r_w} &= 0 \\ \left( -rD \frac{\partial A}{\partial r} + rvS \right)_{r=r_w} &= \frac{Q_w A_0}{2\pi\theta} \\ S(r, 0) &= S_0 \\ A(r, 0) &= 0 \\ M(r, 0) &= M_0 \end{aligned} \quad (6)$$

where  $r_w$  is the radius of the injection well. The downstream boundary of the model aquifer (outlet boundary)  $r_L$  is placed far enough from the injection well so that the solute fronts do not reach the boundary.

To understand the interactive effects of the model parameters upon the contaminant removal rate in a more comprehensive manner, we nondimensionalize Eqs. (1)–(3) by introducing the following transformations:

$$r^* = \frac{r}{\alpha_L}, t^* = \frac{Q_w t}{2\pi\theta\alpha_L^2}, S^* = \frac{S}{S_0}, A^* = \frac{A}{A_0}, \text{ and } M^* = \frac{M}{M_0} \quad (7)$$

The resulting dimensionless equations become

$$R_d \frac{\partial S^*}{\partial t^*} + \frac{1}{r^*} \left[ -\frac{\partial^2 S^*}{\partial r^{*2}} - a_m \frac{\partial}{\partial r^*} \left( r^* \frac{\partial S^*}{\partial r^*} \right) + \frac{\partial S^*}{\partial r^*} \right] = -a_1 f_{BD}^* \quad (8)$$

$$\frac{\partial A^*}{\partial t^*} + \frac{1}{r^*} \left[ -\frac{\partial^2 A^*}{\partial r^{*2}} - a_m \frac{\partial}{\partial r^*} \left( r^* \frac{\partial A^*}{\partial r^*} \right) + \frac{\partial A^*}{\partial r^*} \right] = -a_1 a_2 f_{BD}^* \quad (9)$$

$$\frac{\partial M^*}{\partial t^*} = a_3 f_{BD}^* - a_4 (M^* - 1) \quad (10)$$

$$f_{BD}^* = M^* \left( \frac{S^*}{K_S^* + S^*} \right) \left( \frac{A^*}{K_A^* + A^*} \right) \quad (11)$$

where

$$a_1 = \frac{2\pi\theta\alpha_L^2 q_m M_0}{Q_w S_0}, a_2 = \frac{FS_0}{A_0}, a_3 = \frac{2\pi\theta\alpha_L^2 Yq_m}{Q_w}, a_4 = \frac{2\pi\theta\alpha_L^2 d}{Q_w}$$

$$a_m = \frac{2\pi\theta D_m}{Q_w}, K_s^* = \frac{K_s}{S_0}, \text{ and } K_A^* = \frac{K_A}{A_0} \quad (12)$$

Among the above dimensionless parameters,  $2\pi\theta\alpha_L^2/Q_w$  represents the transport time scale governed by the longitudinal dispersivity and the injection rate. Since  $S_0/(q_m M_0)$  is the time required to consume the background substrate by the initial biomass with the maximum capability of utilizing the substrate,  $a_1$  indicates the ratio of the time scales for transport and substrate degradation under no nutrient limitation. Similarly, the combined dimensionless parameter  $a_1 a_2$  may be considered as the time scale ratio for transport and acceptor degradation under no nutrient limitation. The parameter  $a_3$  is the ratio of the transport time scale to the minimum microbial growth time scale defined by  $1/(Yq_m)$ , while  $a_4$  is the ratio of the transport time scale to the microbial decay time scale  $1/d$ . The parameter  $a_m$  resembles the inverse form of a Peclet number which reflects the relative dominance of transport by molecular diffusion to advective transport.

The dimensionless equations provide insight into the effect of the microbial parameters and the initial biomass concentration upon the local biodegradation rate; that is, the effect of a change in  $q_m$  is mathematically equivalent to the effect of simultaneous changes in  $M_0$  and  $Y$  because these parameters are associated with  $a_1$  and  $a_3$ , respectively. The nondimensionalization of the equations is advantageous for understanding the relative effects of these input data and to reduce the number of numerical simulations required to check the validity of the analytical substrate removal rate.

The boundary and initial conditions (Eq. (6)) are also nondimensionalized by applying Eq. (7). The results become

$$\left( -\frac{\partial S^*}{\partial r^*} - a_m r^* \frac{\partial S^*}{\partial r^*} + S^* \right)_{r^*=r_w^*} = 0$$

$$\left( -\frac{\partial A^*}{\partial r^*} - a_m r^* \frac{\partial A^*}{\partial r^*} + A^* \right)_{r^*=r_w^*} = 1 \quad (13)$$

$$S^*(r^*, 0) = 1$$

$$A^*(r^*, 0) = 0$$

$$M^*(r^*, 0) = 1$$

where  $r_w^* = r_w/\alpha_L$ .

### 3. Theoretical results

For the one-dimensional Cartesian-coordinate bioremediation model, the solute fronts form travelling waves if  $R_d > 1$  (Murray and Xin, 1996; Oya and Valocchi, 1997). This

behaviour enables one to use the travelling wave theory which significantly simplifies the mathematical derivation of the substrate removal rate in the soil column. For the axisymmetric bioremediation model, however, the solute fronts do not have a time-invariant travel velocity, and thus the travelling wave theory used in the Cartesian model is not applicable. In this section we present another approach to derive the analytical substrate removal rate.

We consider mass balance of the substrate and the acceptor in a circular domain of the model aquifer with unit thickness and radius  $r_L^*$  ( $= r_L/\alpha_L$ ) around the injection well. The mass balance equations over the circular domain are obtained by integrating Eqs. (8) and (9) subject to Eq. (13), resulting in

$$2\pi\theta R_d \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} S^* r^* dr^* = -2\pi\theta - 2\pi\theta a_1 \int_{r_w^*}^{r_L^*} f_{BD}^* r^* dr^* \quad (14)$$

$$2\pi\theta \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} A^* r^* dr^* = 2\pi\theta - 2\pi\theta a_1 a_2 \int_{r_w^*}^{r_L^*} f_{BD}^* r^* dr^* \quad (15)$$

Eq. (14) is an integrated mass balance for the substrate; it states that the time rate of change of the total (aqueous and sorbed) substrate in the domain equals the mass flux leaving the domain minus the total mass degraded. The left-hand side of Eq. (14) is the dimensionless displacement rate of the total substrate mass which is negative because the zone containing the substrate reduces in size. Similarly, Eq. (15) is an integrated mass balance for the acceptor; the left-hand side is the dimensionless displacement rate of the acceptor mass. The first term on the right-hand sides of Eqs. (14) and (15) are the dimensionless mass fluxes of the substrate flowing out of the domain and the acceptor supplied from the well, respectively, while the last terms are the removal rates of the substrate and the acceptor, respectively, due to biodegradation. Subtracting Eq. (15) from Eq. (14) multiplied by  $a_2$ , we obtain

$$-a_2 R_d \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} S^* r^* dr^* + \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} A^* r^* dr^* = a_2 + 1 \quad (16)$$

Eq. (16) provides a relationship between the displacement rates for the substrate and acceptor.

For the axisymmetric model, we define the front position by means of the bulk volume displaced by the solute (bulk aquifer volume containing the solute). Fig. 2 illustrates this definition. The front locations are thus mathematically defined as

$$\pi(r_L^{*2} - r_S^{*2}) = 2\pi \int_{r_w^*}^{r_L^*} S^* r^* dr^* \quad (17)$$

for the substrate  $r_S^*$ , and

$$\pi(r_A^{*2} - r_w^{*2}) = 2\pi \int_{r_w^*}^{r_A^*} A^* r^* dr^* \quad (18)$$

for the acceptor  $r_A^*$ . The right-hand sides of Eqs. (17) and (18) represent the bulk volume displaced by the substrate and acceptor, respectively. The above definition of the

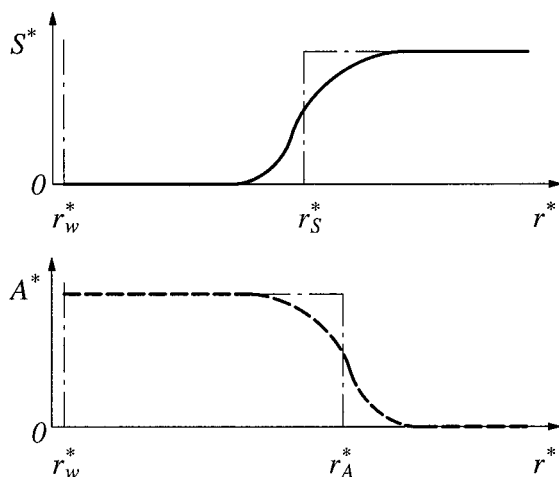


Fig. 2. Definition of the front location.

front location in fact gives the mean travel distances of the fronts from the center of the injection well. Summation of Eqs. (17) and (18) followed by differentiation with respect to  $t^*$  yields

$$\pi \frac{d}{dt^*} (r_A^{*2} - r_S^{*2}) = 2\pi \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} S^* r^* dr^* + 2\pi \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} A^* r^* dr^* \quad (19)$$

Oya and Valocchi (1997) observed in the analysis for the Cartesian bioremediation model that the two fronts travelled in unison. This happens due to the ‘feedback’ effects of the biodegradation and mixing processes. Generation of a mixing zone by dispersion and the velocity difference between the two fronts causes biodegradation to occur in the mixing zone. Since biodegradation reduces the solute concentrations, the fronts become sharper. This sharpening process inhibits further overlapping of the fronts and contracts the mixing zone. Consequently, the biodegradation rate decreases, which then leads to an increase in the extent of the mixing zone. Therefore, the biodegradation and mixing processes interact with each other such that the two fronts neither extensively overlap nor separate from each other. The fronts rather travel downstream together. Similar coupled transport of the solutes is expected to occur in the axisymmetric model. This suggests that  $dr_S^*/dt^* \approx dr_A^*/dt^*$  during the remedial operation which eventually leads to  $dr_S^{*2}/dt^* \approx dr_A^{*2}/dt^*$  in Eq. (19). A key relationship between the rates of change of the displaced bulk volume for the two solutes is thus given as

$$\Delta^* \equiv -2\pi \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} S^* r^* dr^* = 2\pi \frac{d}{dt^*} \int_{r_w^*}^{r_L^*} A^* r^* dr^* \quad (20)$$

Substituting Eq. (20) into Eq. (16), we obtain the approximate dimensionless time rate of change of the displaced bulk volume

$$\Delta^* = \frac{2\pi(a_2 + 1)}{a_2 R_d + 1} \quad (21)$$



Eq. (21) clearly shows that  $\Delta^*$  is constant and dependent only upon the initial and boundary concentrations of the solutes, the stoichiometry of the acceptor consumption, and the retardation factor of the substrate.

The dimensionless rate of substrate mass removed from the model domain  $R_S^*$  is given by the last term of Eq. (14). Substituting Eqs. (20) and (21) into the left-hand side of Eq. (14), we obtain

$$R_S^* = 2\pi\theta a_1 \int_{r_w^*}^{r_d^*} f_{BD}^* r^* dr^* = \frac{2\pi\theta(R_d - 1)}{a_2 R_d + 1} \quad (22)$$

The same procedure for the dimensional model yields the dimensional removal rate  $R_S$  which has a relationship with  $R_S^*$  such that  $R_S = Q_w S_0 R_S^* / (2\pi\theta)$ . This indicates that the analytical substrate removal rate is a function of the background and supplied concentrations of the solutes, retardation factor of the substrate, stoichiometric coefficient for acceptor consumption, and injection flow rate. The rate does not depend upon the dispersion parameters, the microbial kinetic parameters, and the initial biomass concentration. These properties of the analytical substrate removal rate are the same as those for the Cartesian model (Oya and Valocchi, 1997). Disappearance of the microbial kinetic parameters results from the fact that the reaction terms in the substrate and acceptor have the same forms except that the term in the acceptor equation is multiplied with a constant for biodegradation stoichiometry. This enables us to combine the two solute equations by eliminating the reaction terms even after the integration over the radial domain to obtain Eqs. (14) and (15). The insensitivity of the microbial kinetics is significant because it may reduce practitioners' concerns about the impact of the variability of kinetic parameters obtained in experiments upon the long-term efficiency of bioremediation. Furthermore, it should be kept in mind that  $R_S^*$  increases as  $R_d$  increases or  $a_2$  decreases.

The travel velocity of the front is defined as  $dr_S^*/dt^*$  for the substrate and  $dr_A^*/dt^*$  for the acceptor. As stated previously, these velocities are expected to be identical. Neglecting  $r_w^*$ , which is small relative to the model domain, the location of the acceptor front  $r_A^*$  is obtained from the relationship  $\pi r_A^{*2} = \Delta^* t^*$  since  $\Delta^*$  is constant according to Eq. (21). Hence, the theoretical time-variant front velocity  $u_r^*$  for the two solutes is given as

$$u_r^* = \frac{dr_A^*}{dt^*} = \frac{\Delta^*}{2\pi r_A^*} = \frac{1}{2} \sqrt{\frac{\Delta^*}{\pi t^*}} = \sqrt{\frac{(a_2 + 1)}{2t^*(a_2 R_d + 1)}} \quad (23)$$

If the substrate is nondegradable, its front velocity is given as  $u_{nS}^* = 1/\sqrt{2t^* R_d}$  based upon the definition of the front location Eq. (17) with neglecting  $r_w^{*2}$ . This yields the ratio of the theoretical front velocity of the degradable substrate  $u_r^*$  to that of the nondegradable substrate

$$\frac{u_r^*}{u_{nS}^*} = \sqrt{\frac{R_d(a_2 + 1)}{a_2 R_d + 1}} \quad (24)$$

which is constant. Similarly, the front velocity of a nondegradable acceptor is given as  $u_{nA}^* = 1/\sqrt{2t^*}$  which leads to a constant velocity ratio

$$\frac{u_r^*}{u_{nA}^*} = \sqrt{\frac{a_2 + 1}{a_2 R_d + 1}} \quad (25)$$

Since  $R_d > 1$ ,  $u_r^*/u_{nS}^* > 1$  and  $u_r^*/u_{nA}^* < 1$ . Eqs. (24) and (25) indicate that the theoretical front is located between the imaginary nondegradable substrate and acceptor fronts and travels at the velocity which is linearly proportional to the travel velocities of the nondegradable solutes.

#### 4. Numerical results and discussion

We perform numerical simulations to examine the front behaviour and to verify the analytical equation for the substrate removal rate Eq. (22). A numerical model was developed to accomplish these purposes. We used the Galerkin finite element method (FEM) with linear basis functions and the Crank–Nicolson temporal approximation for solving the transport equations (see e.g., Istok, 1989). The nonlinear reaction terms are linearized by taking the Taylor series expansion up to the first-order derivative terms only with respect to the dependent concentration variable to be computed in the equation of interest. The cross-derivative terms with respect to other concentration variables are neglected in order to reduce the size of global matrices. The biomass equation is solved at each node by using the same linearization technique for the nonlinear microbial growth term. The solution for the current time level is obtained by evaluating the zeroth-order terms and the derivatives of the first-order terms with the solution for the previous iteration and incorporating these into the transport component. If the updated solutions for all constituents converge within prescribed error tolerances, the iterative computations for the current time level are terminated. Otherwise the same procedures are repeated by replacing the solutions for the previous iteration with the updated solutions. This iterative numerical technique using the first-order Taylor series expansion may be called a sequential iterative approach with the first-order approximation (SIA-1). The accuracy of the numerical solution was examined for acceptor transport under the assumptions of  $K_S^* \ll 1$ ,  $K_A^* \gg 1$ , and constant biomass that reduce the nonlinear biodegradation term to a first-order decay term and separate the acceptor equation from the substrate and biomass equations. The numerical solution to the resulting equation with arbitrary input data was compared with the analytical solution presented by Tang and Babu (1979). The visual inspection of the results did not detect unacceptable error in the numerical solution.

To observe the front behaviour during bioremediation, we simulate an example case in which groundwater contaminated by toluene is remediated by injecting oxygen. Table 1 summarizes the literature data of the reaction parameters and conditions for aerobic biodegradation of toluene. Note that the biological parameters are strongly dependent upon microbial species and the growth environment (Kelly et al., 1996). For the example

Table 1

Selection of the input data for the biological reaction parameters and the initial conditions used in the example simulation (aerobic biodegradation of toluene)

Parameter	Literature data	Selected data	References
$q_m$ ( $g_S/g_M/day$ )	$1.2 \times 10^{-3}$ –12.5	0.5	Robertson and Button (1987), Kelly et al. (1996)
$Y$ ( $g_M/g_S$ )	0.29–0.5	0.395	Chang and Alvarez-Cohen (1995), Chen et al. (1992)
$d$ ( $day^{-1}$ )	0.048–0.6	0.1	Lawrence and McCarty (1970), Arcangeli and Arvin (1995)
$F$ ( $g_A/g_S$ )	2.19–3.13	2.66	Chen et al. (1992), MacQuarrie et al. (1990)
$K_S$ ( $g_S/m^3$ )	0.01–20.0	0.5	Robertson and Button (1987), Goldsmith and Balderson (1988)
$K_A$ ( $g_A/m^3$ )	$3.52 \times 10^{-4}$ –0.114	0.08	Longmuir (1954)
$S_0$ ( $g_S/m^3$ )	0.1–16.9	5.0	Rittmann et al. (1994), Gersberg et al. (1995)
$M_0$ ( $g_M/m^3$ )	0.046–1.85 <sup>a</sup>	0.427	

<sup>a</sup>The data range of  $M_0$  is based upon the total cell number  $1 \times 10^6$ – $4.04 \times 10^7$  cells/g dry soil (Borden et al., 1986; Harvey et al., 1984), the ratio of active cells to the total cells 0.01 (Staps, 1989), the cell density  $1 \times 10^{-12}$  g/dry cell (Bouwer and McCarty, 1984), the hypothetical bulk soil density  $1.6 \times 10^6$  g dry soil/ $m^3$ , and  $\theta = 0.35$ .

simulation, we selected a set of data from Table 1 (selected data) and computed the dimensionless parameters which are listed in Table 2. Note that we assumed  $\theta = 0.35$ ,  $\alpha_L = 0.02$  m,  $Q_w = 0.5$   $m^2/day$ , and  $A_0 = 8.0$   $g/m^3$  and adopted  $R_d = 1.64$  and  $D_m = 7.34 \times 10^{-5}$   $m^2/day$  from Chen et al. (1992).

Fig. 3 shows the concentration profiles obtained by the example simulation using these input data. Note that the profiles at different observation times are superimposed and plotted on the same radial coordinate. The figure clearly indicates that both the substrate and acceptor fronts travel downstream in unison (Fig. 3a) and create a relatively small overlapped region where microorganisms substantially grow (Fig. 3b). Because of the radial flow field, the front velocity decreases with time. Appearance of this coupled front migration suggests the adequacy of the relationship  $dr_S^*/dt^* \simeq dr_A^*/dt^*$  assumed in the theoretical derivation of the substrate removal rate.

Table 2

Dimensionless input data computed from the data values selected in Table 1

Dimensionless parameter	Data
$a_1$	$7.512 \times 10^{-5}$
$a_2$	1.663
$a_3$	$3.475 \times 10^{-4}$
$a_4$	$1.759 \times 10^{-4}$
$a_m$	$3.228 \times 10^{-4}$
$R_d$	1.64
$K_S^*$	0.5
$K_A^*$	0.01

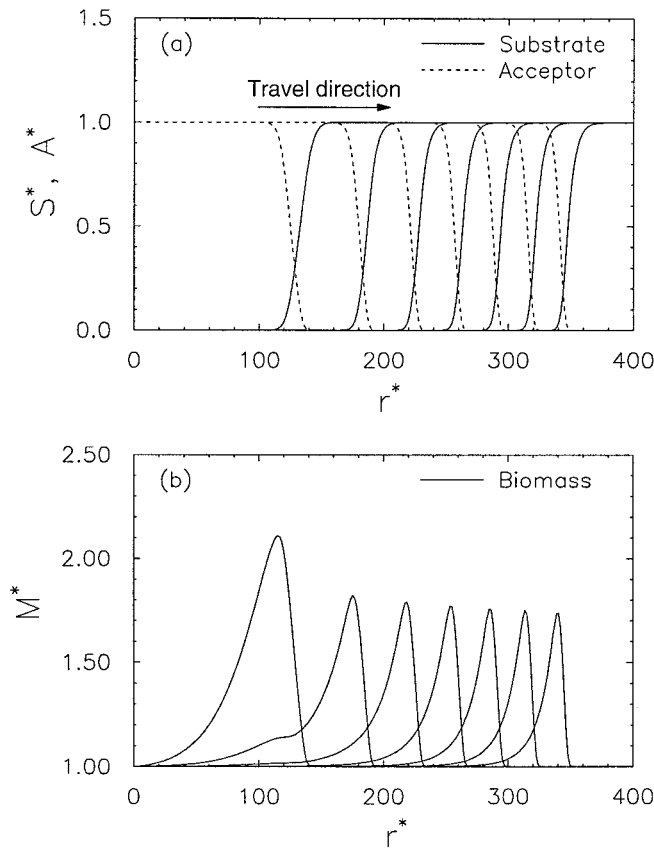


Fig. 3. Concentration profiles at an dimensionless time interval of  $1.2 \times 10^4$  superimposed on the same radial coordinate for the example simulation (input data in Table 2).

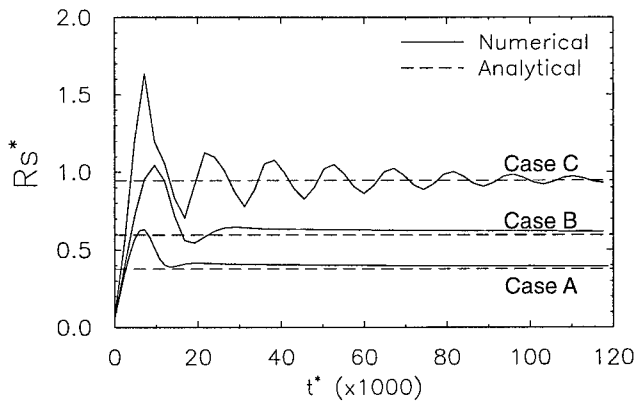


Fig. 4. Comparisons of the removal rate between the analytical ( $R_s^*$ ) and numerical results for  $a_2 = 1.663$  and  $R_d = 1.64$  (Case A, same simulation as Fig. 3),  $a_2 = 0.831$  and  $R_d = 1.64$  (Case B), and  $a_2 = 1.663$  and  $R_d = 5.0$  (Case C). Other input parameters are the same as listed in Table 2.

Fig. 4 compares the analytical removal rates  $R_S^*$  with the numerical removal rates for the example simulation (Case A) and additional two simulation cases in which either  $a_2$  is halved (Case B) by taking a half of the  $F$  value used in Case A (Table 2), and  $R_d$  is increased to 5.0 (Case C). Both additional cases have higher analytical substrate removal rates than Case A (see Eq. (22)). For all cases, the analytical rates are consistent with the numerical results at a sufficiently large  $t^*$ . This indicates that the analytical removal rate given by Eq. (22) is a long-term property of the system.

The physical meanings of the changes in  $a_2$  and  $R_d$  can account for the changes of the removal rate in Cases B and C from that in Case A. Reducing  $F$  in Case B causes a system to have more efficient microbial utilization of the acceptor (lower consumption of the acceptor) and allows the microorganisms to degrade more substrate when the mass flux of the acceptor supplied from the upstream boundary is the same as in Case A. Alternatively, a reduction in  $a_2$  may be obtained by increasing  $A_0$  or by decreasing  $S_0$ . However, these ways also change the  $K_S^*$  and  $K_A^*$  values which affect the microbial capability for solute uptake. In particular, reducing  $a_2$  by taking a smaller  $S_0$  value simultaneously increases  $K_S^*$ ; this may cause the substrate concentration to reach a nutrient-limiting level. We shall later discuss the effect of nutrient-limiting conditions upon the substrate removal rate. In Case C, more substrate mass is initially stored in its sorbed phase; this means that more substrate is available for microbial uptake after instantaneous desorption. Since migration of the substrate plume is significantly retarded under this sorption condition, a larger difference in travel velocity of the fronts results and thus advective mixing of the two solutes becomes more evident. This enhanced advective mixing in conjunction with the higher availability of the substrate yields the higher substrate removal rate.

It is notable in Case C that an interesting oscillatory behaviour appears in the numerical removal rate. Since simulations with much smaller time steps show the same oscillatory phenomenon, the oscillations are not attributed to the numerical method. Oya and Valocchi (1997) analyzed similar phenomena in their Cartesian bioremediation model and showed that the primary factors to generate the oscillations are the interactive feedback effects between biodegradation and advective mixing of the two solutes. In Case C, advective mixing is substantially enhanced by the strong sorption of the substrate (large  $R_d$  value). This significantly enhances the biodegradation process which reduces the solute concentrations in the mixing zone. Since the biodegradation rate decreases as the mixing zone contracts, the mixing process in turn becomes predominant in the system and the cycle repeats. On average, however, the oscillation does not affect the validity of the analytical approximation of the removal rate.

The transport behaviour of the solute fronts is viewed by plotting the time rates of change of the displaced bulk solute volume. In the simulations, the rates (actual rates) for the substrate  $\delta_S^*$  and acceptor  $\delta_A^*$  are obtained by the numerical computations of the second and third terms in Eq. (20), respectively. To show the differences between the actual rates and the theoretical rate  $\Delta^*$  clearly, we plot the relative displacement rates  $\Delta^* - \delta_S^*$  and  $\Delta^* - \delta_A^*$  in Fig. 5. Fig. 5a shows the result for Case A which does not evidently generate front oscillation. In this case, the actual displacement rates for both solutes gradually approach the theoretical rate. The figure indicates that the substrate plume travels slightly faster than the theoretical front, while the acceptor plume travels

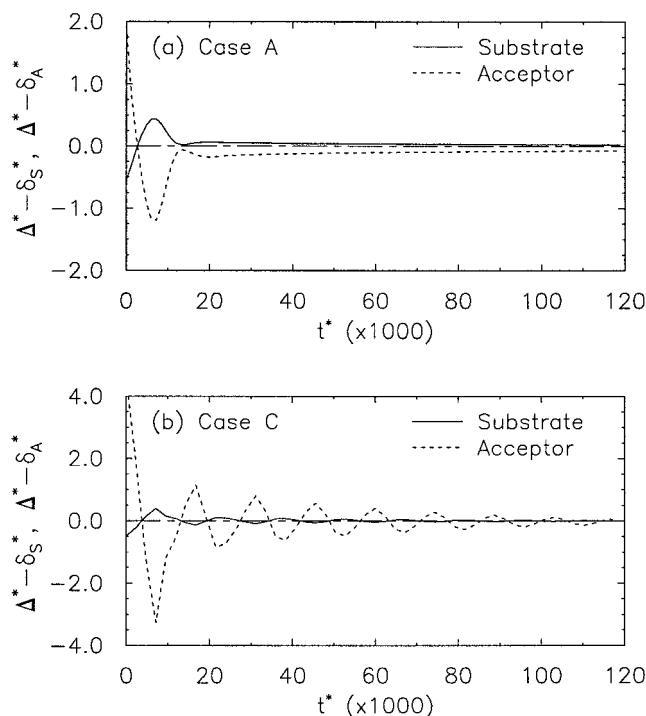


Fig. 5. Relative displacement rates of the substrate  $\Delta^* - \delta_S^*$  and the acceptor  $\Delta^* - \delta_A^*$  for (a) Case A and (b) Case C.

slightly slower than the theoretical front after  $t^* \geq 20 \times 10^3$  ( $t \geq 35$  days). Interestingly, the two relative rate curves never lie in the same quadrant; they barely intersect with or eventually meet each other at a relative rate of zero. This phenomenon is more obvious in Case C which generates damped front oscillations (Fig. 5b). The displacement curves fluctuate about the theoretical rate and cross each other only at rates equal to the theoretical displacement rate. The oscillations die out such that the actual displacement rates finally converge to the theoretical rate. These displacement rate characteristics indicate that the relationship  $dr_S^*/dt^* \approx dr_A^*/dt^*$  leads to an equilibrium state between the biodegradation and mixing processes. Therefore, the relationship is an appropriate assumption for analytical derivation of the long-term substrate removal rate and the front velocity representative for the two solutes in this bioremediation system.

Oya and Valocchi (1997) noted the effects of the initial and boundary solute concentrations and the microbial kinetic parameters upon the short-term behaviour of the substrate removal rate in their numerical simulations of the one-dimensional Cartesian model. For the analytical result for the substrate removal rate to become valid in a relatively short elapsed time, the background substrate and the supplied acceptor concentrations should not be at nutrient-limiting levels for microbial utilization, and sufficient biomass must quickly appear to degrade the solutes effectively after injection of the acceptor begins. In dimensional notation, the unfavourable conditions for the

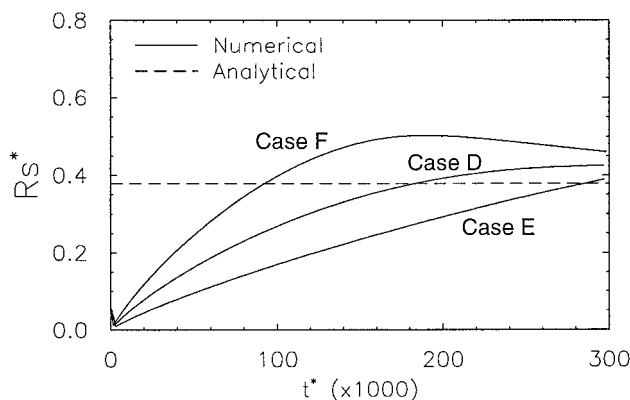


Fig. 6. Comparison of the analytical removal rate with the numerical rates for the cases with increased  $K_S^*$  (Case D), reduced  $a_1$  and  $a_3$  (Case E), and reduced  $a_1$  along with increased  $a_4$  (Case F). Other parameter values are the same as those for Case A (Table 2).

application of the analytical method to the short-term removal rate may be represented by (a) a large  $K_S$  or  $K_A$  value compared with  $S_0$  or  $A_0$ , (b) a relatively small  $q_m$  value, or (c) a small  $Y$  value or a large  $d$  value along with a small  $M_0$  value. Condition (a) causes the initial substrate or the supplied acceptor to be subject to a nutrient-limiting condition. Condition (b) indicates a potentially negligible microbial capability of degrading the substrate and acceptor. Condition (c) results in a low biomass concentration due to no effective cell growth and an insufficient initial microbial population. Conditions (b) and (c) actually mimic a system having nondegradable substrate and acceptor which is not practically relevant for bioremediation studies. In dimensionless notation for the axisymmetric model, condition (a) is equivalent to  $K_S^* > 1$  or  $K_A^* > 1$ , while conditions (b) and (c) are produced by decreasing  $a_1$  with a concomitant decrease in  $a_3$  or a concomitant increase in  $a_4$ .

Fig. 6 plots the numerical results for the substrate removal rate for cases in which  $K_S^*$  is increased to 25 (Case D), both  $a_1$  and  $a_3$  are changed by a factor of 0.02 (Case E), and  $a_1$  and  $a_4$  are changed by the respective factors of 0.04 and 5 (Case F). These parameter changes are all relative to Case A. The plots are compared with the analytical removal rate which is unchanged from Case A ( $R_S^* = 0.378$ ). The numerical results of all cases show substantially prolonged elapsed times to attain the removal rate equal to the analytical value compared with Case A (see Fig. 4). This indicates that the analytical method overestimates the removal rate for a short-term prediction under conditions of nutrient limitation or when there are inefficient microbial growth kinetics with the low initial biomass concentration. The removal rates have peaks and eventually decrease toward the analytical rate (data not shown) as seen in Case A. Additional simulations with various  $K_S^*$  and  $K_A^*$  values showed that the analytical method is inappropriate for short-term predictions of the removal rate when  $K_S^* \gg 1$  or  $K_A^* \gg 1$ . This finding is consistent with that for the Cartesian model (Oya and Valocchi, 1997). To find the criteria for  $a_1$ ,  $a_3$ , and  $a_4$ , we need further investigations on their quantitative effects upon the short-term behaviour of the removal rate.

## 5. Conclusions

In this paper, we developed an analytical approach to approximate the long-term substrate removal rate for in situ bioremediation of a contaminated porous medium under ideal radial flow conditions. The mathematical derivation involved an assumption that the substrate and acceptor fronts travel in unison away from the injection well. This coupled migration of the solute fronts was expected from the results reported by Oya and Valocchi (1997) who analyzed travelling wave phenomena in a one-dimensional Cartesian-coordinate bioremediation model. The mathematical procedures in this study showed that the long-term rate of substrate mass removal Eq. (22) is independent of the dispersion parameters, microbial kinetic parameters, and initial biomass condition. This long-term rate is instead dependent on the injection flow rate, the background substrate and injected acceptor concentrations, the stoichiometric ratio of the substrate-acceptor reaction, and the retardation factor of the substrate. This finding is consistent with that reported for the Cartesian model (Oya and Valocchi, 1997) and is of practical importance if the bioremediation scenario assumed in this study can represent actual remedial conditions. The insensitivity of the long-term biodegradation rate to the microbial kinetic parameters suggests that it might not be necessary to estimate these parameter values to a high level of accuracy. This may reduce the temporal and financial burdens incurred when conducting detailed experiments for parameter estimation. It is interesting that the long-term removal rate formula is similar to that for the Cartesian model, even though the governing equations for the radial nonuniform case are so much more complex.

The validity of the analytical approach was examined by comparing the analytical removal rate with numerical results for a case representing aerobic biodegradation of toluene. The comparisons proved that the analytical method can adequately yield the long-term removal rate.

The numerical results also showed the existence of an initial transient period during which the substrate removal rate is strongly dependent upon the microbial kinetic parameters and the initial biomass concentration. One important criterion for the applicability of the analytical removal rate to the short-term prediction is that the background and supplied solute concentrations should not be at the nutrient-limiting level. In addition, the microbial kinetic parameters and the initial biomass concentration should be adequate enough so that the microorganisms can efficiently grow on and consume a sufficient amount of the substrate. Since the applicability of the analytical method to the short-term prediction seems practically important, further study of this initial time period is necessary.

The evidence of coupled front transport is a very noteworthy phenomenon which leads to significant mathematical simplification enabling the derivation of the substrate removal rate and front velocity. This study showed that the theoretical approach based upon this front behaviour is a new alternative method to derive the long-term removal rate without transforming the fixed physical coordinate to a moving coordinate according to the travelling wave theory as performed for the Cartesian model by Oya and Valocchi (1997). Therefore a priori confirmation of travelling wave formation is not necessary in the alternative method. Compared with the analytical method based upon the travelling wave theory, this alternative approach is considered to have higher



potential applicability to theoretical analyses of the solute transport and biodegradation characteristics in more complex, realistic bioremediation systems that may involve kinetic sorption/desorption, aquifer heterogeneity, and higher spatial dimensions. We are now applying the new approach to the analysis of more complex systems.

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